

**CONFIDENTIAL INFORMATION  
SUBJECT TO PROTECTIVE ORDER**

**IN THE UNITED STATES DISTRICT COURT  
FOR THE DISTRICT OF COLUMBIA**

ELF ATOCHEM NORTH AMERICA, INC.)

Plaintiff,

v.

Q. TODD DICKINSON,

Commissioner of Patents  
and Trademarks

Defendant.

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Civil Action No. 1:99CV02559(TPJ)

**DECLARATION OF DR. MICHEL FOURE  
SUBMITTED PURSUANT TO FED. R. CIV. P. 56 AND LOCAL RULE 11.2  
IN SUPPORT OF PLAINTIFF'S OPPOSITION TO DEFENDANT'S MOTION FOR  
SUMMARY JUDGMENT**

I, Michel Foure, do hereby declare that:

1. I am a Doctor Rerum Natur in Chemistry; this graduate doctorate degree was conferred on me by the State of Wurzburg (West Germany) in 1975, and is equivalent to a Ph.D. degree here in the United States. I did post-doctorate research in chemistry at the University of Osaka (Japan) from 1975-1977.

2. I was employed at the Lacq, France, Research Centre of Societe Nationale Elf Aquitaine from 1977 to 1980, doing research in polyvinyl chloride ("PVC") stabilizers, cosmetics and fragrances, lubricant, process development, and organotin biocides. In 1980, I took a position at M&T Chemicals Inc. as a Senior Project Chemist in the PVC Additives Group. In 1982, I was

promoted to the position of Manager of the PVC Additives Group. I currently hold the position of Director of Strategic Research for Elf Atochem North America, Inc. ("Atochem"), the successor of M&T Chemicals Inc. As part of my responsibilities over the course of my professional career in industry, I have had direct responsibility for PVC stabilization/additives research programs since 1978. I hold five patents in the field of PVC stabilization. My curriculum vitae is attached as Ex. C.

3. I have reviewed and am familiar with the following materials:

- a. the patent specification for Serial No. 07/870,759 ("the '759 application") [Ex. D];
- b. the pending claims [Ex. E];
- c. the following prior art patents [Exs. F through M, respectively]:

<u>Inventor</u>	<u>U.S. Patent No.</u>	<u>Ex.</u>
Gough et al.	3,928,285	F
Stapfer et al.	3,830,751	G
Hechenbleikner et al.	3,167,527	H
Hechenbleikner et al.	3,196,129	I
Weinberg et al.	2,832,750	J
Kauder et al.	3,817,915	K
Wowk	3,758,537	L
Schroeder	3,595,893	M

d. the February 23, 1999, decision of the Board of Patent Appeals and Interferences ("Board") [Ex. N]; and the Board's July 27, 1999, decision on Request for Rehearing [Ex. O].

e. my Declaration, dated August 18, 1982, which I understand was filed with the U.S. Patent and Trademark Office in connection with a patent application that is related to S.N. 07/870,759 [Ex. P];

f. the results of testing performed by Dr. Christopher Bertelo, comparing the claimed invention with a stabilizer system disclosed (but not tested) in the Gough patent [Ex. Q].

In addition, based on my education, training, and experience, I am familiar with the state of the art as of the late 1970s pertaining to polyvinyl halide ("PVH") stabilization, including PVC stabilization, which is one important aspect of PVH stabilization; and

g. the Memorandum in Support of Defendant's Motion for Summary Judgment ("Defendant's Memo").

#### **I. The Field of PVH Stabilization and the Chenard Invention**

4. Atochem is a leading chemical manufacturer, whose headquarters are located in Philadelphia, Pennsylvania. Atochem develops and sells chemical products that aid in the manufacture of certain PVH polymers. The class of PVH polymers includes PVC--polyvinyl chloride--a plastic material that is commonly used in everyday products such as plastic pipe, siding, injection-molded parts, and plastic bottles.

5. PVH products are generally manufactured from molten PVH by an extrusion process. The manufacturing process requires the application of heat, which unfortunately can chemically degrade the PVH. This degradation is manifested by discoloration and increased viscosity, and can lead to performance defects such as brittleness and diminished resiliency, durability, and strength.

6. In order to counteract these problems, workers add certain chemicals, called "stabilizers," to the PVH material during manufacturing. Stabilizers inhibit the chemical degradation and thus prevent the discoloration from white to black that would normally occur in the absence of stabilizers. Stabilizers are essential for the manufacture of usable, high-quality PVH products.

7. Researchers have long concentrated their efforts on improving PVH stabilizers. Many, but not all, of these stabilizers contain tin as the main ingredient, which is very expensive in the quantities necessary for effective stabilization.

8. In the late 1970s, Drs. Chenard and Mendelsohn – researchers at Societe National Elf Aquitaine in Lacq, France, invented a stabilizer combination which significantly reduced the amount of tin required to achieve acceptable stabilization, thereby providing substantial savings. In my opinion, their invention (“the Chenard invention”) was a significant advance in the field of PVH stabilization.

9. Researchers had been working to develop and improve PVH stabilizers long before Drs. Chenard and Mendelsohn invented their stabilization system. In general, such researchers had either a graduate or undergraduate degree in chemistry coupled with some experience in the field of plastics manufacturing.

## **II. The Chemistry of the Chenard Invention**

10. The invention by Drs. Chenard and Mendelsohn relates to a composition comprising (1) a monoorganotin or diorganotin compound in which at least one tin atom is bonded to a sulfur or a halogen atom (“the claimed organotin compounds”) and (2) a mercapto alkanol ester of a monocarboxylic acid (“the claimed reverse ester”) (collectively hereafter, “the claimed composition”). The Chenard invention also relates to methods of stabilizing a “vinyl halide resin,” or PVH polymer, using the claimed composition.

11. As the name implies, organotin compounds are organic compounds<sup>1</sup> that contain atoms of the metallic element tin. The organotin class of chemical compounds is extremely large. Not all organotin compounds, however, fall within the scope of the claimed compositions.

12. There are two specific types, or subclasses, of organotin compounds that fall within the scope of the claimed compositions; they differ from one another based on the types of chemical atoms they contain in addition to the tin. Tin generally can have four atoms bonded (directly attached) to it. Among the many different atoms that can bond to tin are carbon, sulfur, oxygen, or halogens.<sup>2</sup> The first type of claimed organotin compound, called the "claimed organotin-sulfur compound," must have a tin atom bonded to a sulfur atom and to one or two organic groups (which contain one or more carbon atoms). I understand that the Defendant calls the claimed organotin-sulfur compound "Compound A." The second type of claimed organotin compound, called the "claimed organotin-halide compound," must have a tin atom that is bonded to a halogen atom and to one or two organic groups. I understand that the Defendant calls the claimed organotin-halide compound "Compound A' (A-prime)."

13. The second required component of the claimed composition is the claimed reverse ester. Its chemical structure is represented by the chemical formula<sup>3</sup>  $R^1-CO_2-R^2-SH$ . The claimed

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<sup>1</sup> The broad system of chemicals known as organic compounds is based on the presence of one or more carbon atoms attached to various other atoms.

<sup>2</sup> Halogen is the generic term for the group of chemical elements consisting of fluorine, chlorine, bromine, iodine, and astatine. These various halogens have similar chemical and physical characteristics. Halides are chemicals containing halogens. Hence, polyvinyl chloride is polyvinyl halide containing the halogen chlorine.

<sup>3</sup> In the chemical formulas appearing in this Declaration, "R," "R<sup>1</sup>" and "R<sup>2</sup>" represent various types of organic groups, "O" represents oxygen, "S" represents sulfur, "H" represents hydrogen, "C" represents carbon, "Sn" represents tin, "B" represents boron, and "N" represents

reverse esters are a very small subclass of the larger class of reverse esters, which in turn are a small subclass of the *very* large class of sulfur-containing organic compounds.

14. The chemical structures and complexity of sulfur-containing organic compounds vary enormously. For example, the chemical classes known as mercapto acid esters ( $R^1-O_2C-R^2-SH$ ), simple thiols ( $R-S-H$ ), sulfides ( $R^1-S-R^2$ ), disulfides ( $R^1-S-S-R^2$ ), mono thioanhydrides ( $R^1-CO-S-CO-R^2$ ), dithioanhydrides ( $R^1-CS-O-CS-R^2$ ), trithioanhydrides ( $R^1-CS-S-CS-R^2$ ), sulfoxides ( $R^1-S(O)-R^2$ ), sulfones ( $R^1-SO_2-R^2$ ), thioketones ( $R^1-C(S)-R^2$ ), sulfates ( $R^1-O-SO_2-O-R^2$ ), thioesters ( $R^1-C(S)O-R^2$ ), dithioesters ( $R^1-C(S)S-R^2$ ), monothiocarbonates ( $R^1-SCO_2-R^2$ ), dithiocarbonates ( $R^1-SC(S)O-R^2$ ), trithiocarbonates ( $R^1-SCS_2-R^2$ ), thioureas ( $R^1R^2N-C(S)-NR^3R^4$ ), and sulfonate esters ( $R^1-SO_3-R^2$ ) are all sulfur-containing organic compounds. Sulfur-containing organic compounds may also contain countless other functional groups not listed here.

15. Sulfur-containing compounds in general all differ from each other in the number and type of atoms present and in the way the atoms are connected to each other. Each genus or subclass of compounds, as well as specific compounds within each genus or subclass, exhibits unique chemical and physical properties. Additionally, there are theoretically infinite numbers of compounds which fall within each genus or subclass of sulfur-containing compounds.

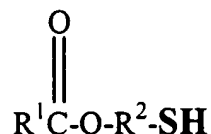
16. The claimed "reverse esters" are so named because as compared to the class of compounds known as "mercapto acid esters," the position of the sulfur part of the molecule, called the mercapto (also called "thiol" or "-SH") group, relative to another part called the ester ("CO<sub>2</sub>-")

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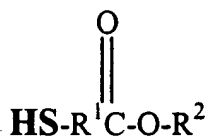
nitrogen. The subscripts indicate how many atoms of a particular type are present; hence -CO<sub>2</sub>- indicates a functional group with one carbon atom and two oxygen atoms. The superscripts serve to distinguish chemical groups from one another. Thus R<sup>1</sup> represents an organic group that is distinct from R<sup>1</sup>. Note that R<sup>1</sup> and R<sup>2</sup> may be the same or different from one another.

group,<sup>4</sup> is reversed. The reverse ester differs from a mercapto acid ester as illustrated by the following structures:

mercapto alkanol ester (reverse ester):



mercapto acid ester:

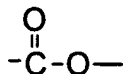


17. Thus, in the reverse ester, the -SH group is attached to the "R<sup>2</sup>" group, whereas in the mercapto acid ester, the -SH group is attached to the "R<sup>1</sup>" group. This difference in position makes a surprisingly remarkable difference in terms of stabilization activity.

18. The claimed reverse esters themselves form a small subclass of reverse esters, especially as they are further defined in the claims as mercapto alkanol esters "of a monocarboxylic acid." This means that the compound contains only one ester or (-CO<sub>2</sub>-) group.

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<sup>4</sup> The -CO<sub>2</sub>- group is also denoted by chemists as follows:



Single lines denote what chemists call "single bonds;" double lines denote "double bonds." Thus the -CO<sub>2</sub>- ester group, depicted above, has one oxygen atom bonded to the carbon atom through a double bond, and the other oxygen through a single bond. This very specific configuration of carbon and oxygen atoms is common to all ester groups.

### **III. The Prior Art References**

19. In my opinion, as explained below, none of the cited prior art references, considered separately or in combination, disclose, teach or suggest the claimed invention.

#### **The Gough Patent**

20. I understand that the Board relied primarily on the Gough patent [Ex. F] to reject the claims to the Chenard invention as obvious. In my opinion, the Board misinterpreted the Gough patent. In my view, the Gough patent teaches a PVH stabilizer system that researchers in the field of PVH stabilization would have recognized as commercially useless. For this reason, researchers would have discounted Gough's statements regarding the superiority of his stabilizer system. Furthermore, a claimed reverse ester was presented by Gough as but one of a large number of organic thiols that Gough disclosed could be added to organotin-borates as co-stabilizers. The reverse ester was not highlighted or emphasized in any way; its superiority as a co-stabilizer was not even hinted at in the Gough patent. It is my opinion that researchers in the PVH stabilization field during the 1970s would not have known or even guessed from the Gough reference that reverse esters would be effective as co-stabilizers for certain known organotin compounds that Gough allegedly was improving upon. For these reasons, developed more fully below, I disagree with the Board's conclusions that it would have been obvious from Gough to select a claimed reverse ester and combine it with a claimed organotin compound.

21. The Gough patent teaches the alleged unique synergistic combinations of "organotin-borates" with organic thiol compounds (i.e., organic compounds containing a -SH group) as stabilizers for PVC. "Organotin-borates" constitute a very narrow subclass of



organotin compounds that contain, in addition to tin, a chemical "borate" group<sup>4</sup> bonded (attached) to the tin atom. Organotin-borates are entirely different from the claimed organotin compounds.

22. Gough dismisses prior art stabilizers because they had not been entirely satisfactory, were expensive, difficult to make, or had undesirable properties such as being extremely smelly or imparting initial color to the plastic. [Ex. F, col. 3, lines 50-66.] Gough then presents organotin-borate combinations as a solution to these problems.

23. In my opinion, researchers would have viewed the selection of this narrow class of organotin-borates from among the members of the broad organotin compounds as the main focus of the Gough patent.

24. Gough combined the narrow class of organotin-borates with thiol compounds, an extremely broad class of -SH containing compounds. Gough sets forth requirements that the thiol compound have a molecular weight from 40 to 400 per thiol group and a boiling point of at least 180°C at one atmosphere pressure. [Ex. F, col. 2, lines 16-20.] These requirements do not substantially guide the selection of suitable thiol compounds. An enormous number of compounds meet these general requirements.

25. Gough supplies general formulas which would encompass just about every possible thiol compound. Gough provides the reader of his patent with the impression that the use of any thiol compound within these broad general formulas could be used to improve the stabilization performance of his particular, new, narrow class of organotin-borates. Gough thus

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<sup>4</sup> A borate group contains both boron and oxygen in a specified configuration.

cannot be said to point in the direction of the specific, narrow class of reverse esters claimed by Drs. Chenard and Mendelsohn.

26. The Gough patent also lists 29 illustrative examples of thiols, representative of the variety of thiols in the broad classes disclosed by Gough. Only one is a claimed reverse ester. But there is nothing in Gough to suggest that this particular reverse ester would function as an effective co-stabilizer with the claimed organotin compounds. In fact, there are no test results, or any other indication of stabilization properties, pertaining to this particular claimed reverse ester disclosed in the Gough patent.

27. Gough reports data for several of his organotin-borate/thiol combinations, and his purported results are not impressive. Gough measured the amount of time it took to reach a certain level of discoloration. The longer it took to develop color, the better the result in the data reported by Gough. According to cols. 13 and 14, Table 1, Example 7, the control, DMTBOT, (a conventional organotin-sulfur compound within the scope of the claimed invention and used alone) did not develop a gray color until 10 minutes after processing. In contrast to the control, Gough's own organotin-borate/thiol compositions in Examples 14 and 15 are reported as taking even less time (8 and 6 minutes, respectively) to develop significant color (tan or gray), indicating that Gough's combinations provide *less* stabilization than the conventional organotin-sulfur stabilizer alone.

28. In my opinion, researchers in the PVH stabilization field working in the mid to late 1970s would have understood, from the Gough patent disclosure alone, that the Gough stabilizer system was ineffective. Such researchers would have dismissed the Gough stabilizer system as commercially useless. In fact, such researchers would not have given the Gough patent

serious regard because Gough's results undercut his assertions about his contribution to the field of PVH stabilization. [Ex. F, col. 2, lines 13-16.]

29. A comparison of DMTBOT with the Gough organotin-borate alone (*i.e.*, no thiol compound included with either organotin stabilizer) further demonstrates the inferiority of Gough's system. The use of DMTBOT alone at 0.5 phr (parts per hundred parts resin by weight), as mentioned above, took 10 minutes to develop significant color. [Ex. F, cols. 13-14, Table I, Example 7.] This far exceeded the stabilization by the organotin-borate alone at 0.5 phr. (Ex. F, cols. 13-14, Table III, Example 27, which took only 2 minutes to develop significant early color.)

30. The organotin-borate compound has a higher tin concentration than the DMTBOT. Researchers in the PVH stabilization industry understand that tin content is a good indication of stabilization performance, with higher levels of tin generally giving better stabilization. Even with a higher tin content, the Gough organotin-borate alone is far inferior to the organotin-sulfur compound, DMTBOT.

31. Furthermore, the alleged superior results of Gough's organotin-borate/thiol combinations would have been questioned by researchers in the PVH industry because his method for determining stabilization performance was highly subjective. Gough subjectively evaluated the discoloration as various shades of "white," "light cream," "light yellow," "light gray," "gray" and "dark gray" at incremental periods of time. Those in the industry would have understood that many degrees of discoloration exist within those color categories.

32. In my view, had researchers tested the Gough stabilizer combinations, they would have understood from the results that the Gough combinations were unacceptable for commercial

use. This would have confirmed what they would have understood from the Gough patent: that the Gough stabilization system was useless. Such researchers would have immediately rejected the Gough combinations. As discussed in Sections V and VI of this Declaration, further recent testing bears this out: organotin-borates in combination with other sulfur-containing compounds produced terrible results. But even from the Gough patent itself, it was well known as of August 29, 1978, that the organotin-borate/thiol combinations of Gough were of no practical use for stabilizing PVH polymers.

33. For these reasons, I strongly disagree with the Board's conclusion that one of ordinary skill in the art would have understood that the synergistic combinations of Gough were superior to other known combinations of stabilizers. [Ex. N, Board Decision, at page 14.] Quite the opposite is observed from reading the Gough patent, and could easily be confirmed by testing the Gough stabilizers. Gough's organotin-borates--both alone and in combination with thiols--are inferior to other stabilizers.

34. Thus, I also disagree with the Board's position that researchers would have been taught by the Gough patent that thiols should be combined with conventional stabilizers. [See Ex. N, Board Decision, at page 13.] Rather, one would have been taught that thiols as secondary stabilizers cannot improve Gough's special class of compounds to an acceptable level.

35. In the background section, the Gough patent mentions other, known stabilizers, including the specific reference to the combination of an organotin-sulfur compound with a thioanhydride. A thioanhydride is a sulfur-containing compound, but is neither a claimed reverse ester nor a thiol. The Gough patent refers to the prior stabilizers in general, and the organotin-sulfur/thioanhydride combination in particular, as examples of the prior art that Gough was

seeking to improve upon. Gough specifically points out the disadvantages of the known stabilizers which his invention is designed to overcome:

Many of these [prior PVH stabilizers] which have been used or suggested to be used to improve the resistance of [PVH polymers] to early color development during processing *have not been entirely satisfactory, or are expensive or are difficult to make, or have undesirable properties such as being odoriferous during processing of the plastic or in themselves impart some initial color to the plastic.*

[Ex. F, Gough, col. 1, lines 59-66 (emphasis added)]. Thus, these prior stabilizers, including the organotin-sulfur/thioanhydride combination, are examples of what Gough considers inferior to his organotin-borate/thiol combination. Although researchers would have disregarded the Gough organotin-borate/thiol stabilizer system as unacceptable, in my opinion, they would nevertheless have agreed with the above statement as to the shortcomings of the then-existing PVH stabilizers.

36. Moreover, Gough provides no teaching as to why Gough selected thiol compounds to improve the performance of organotin-borates. Gough's teaching about which thiol compounds were useful for the invention consists of (1) very general molecular weight per thiol group and boiling point restrictions which encompass infinite thiol compounds; (2) general formulas which also encompass an almost unlimited number of infinite thiol compounds; (3) a list of 29 compounds with no explanation about why such compounds are "usable"; and (4) the compounds Gough actually tested, which includes those compounds Gough chose to demonstrate his invention, but does not include any reverse esters.

37. Nothing in the Gough patent points in the direction of reverse esters. Although Gough very generally discloses reverse esters and mentions one claimed reverse ester, he only does so in the context of an extremely broad disclosure generically covering thiol compounds

that could be used with the organotin-borates. In my view, as explained below, researchers would have had no reason to select a claimed reverse ester from among the extremely broad class of thiol compounds disclosed in Gough.

38. Gough discloses that suitable organic thiols include those falling within four general formulas (d), (e), (f) and (g), appearing in col. 3, lines 50-69 of the Gough patent. These four general formulas encompass an uncountable number of thiol compounds falling in widely divergent thiol subclasses. Although the claimed reverse esters do fall within the scope of formula (g), they are not highlighted or distinguished in any respect. The claimed reverse ester is thus but one small, apparently unremarkable aspect of the large class of thiols described by Gough.

39. In addition to the general formulas, Gough lists 29 “usable” organic thiol compounds at col. 8, beginning at line 50. Gough provides no teaching about what qualities of these compounds put them as candidates on the “usable” list, other than membership in Gough’s broad general formulas. Only one of these compounds, 2-thioethyl octanoate, is a claimed reverse ester. Again, nothing in Gough suggests that this compound differs in any respect from the enumerated compounds. In fact, the Gough patent does not even suggest that the enumerated compounds differ from those disclosed more generically by formula.

40. As mentioned above, the Gough patent includes test results for some of the organotin-borate/thiol combinations it describes. Gough selected eight thiols for testing

purposes; six were mercapto acid esters<sup>5</sup> and two were simple thiols.<sup>6</sup> None were the claimed reverse esters.

41. Gough did not report test results for 2-thioethyl octanoate. In fact, Gough did not report test results for any reverse esters, much less of any *claimed* reverse esters, in conjunction with any organotin compound. Nor did Gough indicate how 2-thioethyl octanoate--or any of the reverse esters--would perform as stabilizers relative to the other thiols that Gough actually tested. One reading the Gough patent would understand Gough to expect that all of the disclosed thiol compounds are equivalent in performance as co-stabilizers for organotin-borates or at least to expect no better results with reverse esters than other thiols.

42. Researchers would have looked, if to anything, to the compounds Gough actually tested to discern what compounds Gough considered to be most suitable. Gough chose to test only compounds falling within formulas (e) and (f) (simple thiols and mercapto acid esters, respectively, which do not encompass the claimed reverse esters).

43. Researchers would have understood Gough's preferred thiol compounds to be those Gough actually tested. Thus, researchers would have understood Gough to teach that the thiols that work best with the organotin-borates were those that fall within formulas (e) and (f). They would not have interpreted the claimed reverse esters, which do not fall with formulas (e) and (f), to be superior to those actually tested. Gough provides absolutely no teachings about the

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<sup>5</sup> The following mercapto acid esters were tested in combination with Gough's organotin-borates: octylthioglycolate, isooctyl thioglycolate, isooctyl  $\beta$ -mercaptopropionate, octadecyl thioglycolate, n-butyl thioglycolate, and benzyl thioglycolate.

<sup>6</sup> The following simple thiols were tested in combination with Gough's organotin-borates: dodecyl mercaptan and octyl mercaptan.

performance of compounds within formula (g), and provides no motivation to try these compounds in place of the compounds actually tested, much less any of the many other types of compounds that could have been selected from Gough's disclosure.

44. Gough provides no actual test data or any other information that suggests the reverse ester would work any better than Gough's other thiol compounds as a co-stabilizer for organotin-borates. There is no basis in Gough from which researchers could assume that reverse esters would work to enhance the stabilization performance of any other organotin compound, such as a claimed organotin-sulfur or organotin-halogen compound.

45. Contrary to the Board's Decision, researchers prior to August 29, 1978 would *not* have had a reasonable expectation that the combination of the claimed organotin compounds and the claimed reverse esters would successfully function as a commercially acceptable PVH stabilizer. [See Ex. N, Board Decision, at page 15.] Given the enormous number of all possible combinations of tin stabilizers with secondary stabilizers such as sulfur-containing compounds, researchers could not possibly have had a reasonable expectation of success for the claimed invention. Researchers sometimes, but not always, obtained synergism between combinations of stabilizer compounds. Antisynergism was also sometimes observed, however, wherein the addition of a secondary stabilizer actually *decreases* the activity of the primary tin stabilizer. Such antisnergistic combinations presented little promise for improved stabilizers. As of the late 1970s, skilled PVH stabilization researchers had no way to predict such synergism or antisnergism between different types of stabilizer compounds other than by experimenting. This is still true today.



46. Indeed, the claimed reverse esters were not known to work by themselves as stabilizers. [Ex. D, '759 application, at page 25.] Researchers would not have expected them to give synergistic results--or the highly unexpected and superior synergistic results--obtained by Chenard and Mendelsohn when combined with the claimed organotins. I therefore disagree with the Board's statement that one of ordinary skill in the art would have had a reasonable expectation of obtaining a synergistic effect when utilizing organic thiols and organotin mercaptides as stabilizers in vinyl halide polymers. [Ex. O, Board's Decision on Rehearing, at page 3.]

47. Gough provides no motivation to select the claimed reverse ester from among its disclosure of an infinite number of thiol compounds or to combine that ester with a claimed organotin. *At most*, someone in the PVH stabilization industry would have expected a claimed reverse ester to perform equivalently to the other thiol compounds in Gough when combined with an organotin-borate. The claimed invention was not suggested or taught by the Gough patent.

#### **The Stapfer Patent**<sup>7</sup>

48. I understand that the Board also cited the Stapfer patent [Ex. G] against the pending claims. In my opinion, the Board misinterpreted the Stapfer patent. It does not, contrary to the Board's finding, teach or suggest any specific combination of an organotin-sulfur compound and a sulfur-containing compound, much less the claimed combinations. [See Ex. N

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<sup>7</sup> The Stapfer patent currently cited against the application is U.S. Patent No. 3,830,751. I understand that a different Stapfer patent, U.S. Pat. No. 3,890,276 [Ex. S], was also cited by the Examiner and discussed by the applicants during prosecution of the pending 07/870,759 application. It is my opinion, however, that neither Stapfer patent teaches, suggests, or motivates those of skill in the art to arrive at the claimed invention.

Board Decision, at page 14; Ex. O, Board's Decision on Rehearing, at page 3.] Stapfer's broad, vague comment that poor stabilizers can be used in combinations which exhibit synergism provides no guidance to skilled researchers as to which compounds to combine to achieve such "synergism." It certainly does not provide any expectation that any given combination will indeed exhibit such synergism. For the reasons set forth below, it is my opinion that the Stapfer patent has no relevance to the claimed combination. Indeed, as demonstrated below, Stapfer relates to an entirely different type of invention.

49. Stapfer's patent teaches that known PVH stabilizers work more effectively when suspended on a solid support. Thus, Stapfer discloses the suspension of known stabilizers on an inorganic substrate of synthetic silicates. The focus of the Stapfer patent is not on any particular PVH stabilizers or combinations thereof, but rather on the use of the silicate powder to support and enhance the performance of stabilizers.

50. The Stapfer patent lists representative examples of the various types of stabilizers that could be used with the silicate powders:

The following list of stabilizing compounds are suitable for practicing the present invention. While many of the compounds listed are poor stabilizers alone, they may be used in combinations which exhibit synergism that are further improved by the present invention. [Ex. G, col. 3, lines 14-18.]

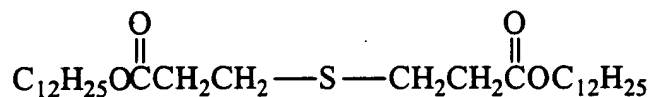
51. The Stapfer patent then lists at least 17 extremely broad classes of PVH stabilizers. Stapfer includes numerous representative examples for each class. [Ex. G, cols. 3-11]. This list covers, both generically by class and specifically by chemical name, an enormous number and variety of stabilizers. The list includes compounds containing tin, sulfur, or both.

52. Only three of these classes are sulfur compounds that do not contain tin.

According to the PTO, "Stapfer also discusses thiocarboxylates, which are organic compounds containing sulfur in the same class as Compound B [reverse esters]." *See* Defendant's Memo at page 15. I disagree. In my opinion, the thiocarboxylate sulfur compounds listed by Stapfer are very different from the claimed reverse esters and would not have been considered similar to the claimed reverse esters by those in the PVH stabilization industry.

53. I also disagree with the Defendant's conclusion that "Stapfer directly discloses that [an organotin-sulfur compound] and a compound similar to [a reverse ester] may be combined to form a PVC stabilizer." *See* Defendant's Memo at page 15. Nowhere does Stapfer "directly disclose" that an organotin-sulfur compound should be combined with any non-tin, sulfur-containing compound, much less any sulfur compound that a skilled artisan would deem "similar" to a reverse ester.

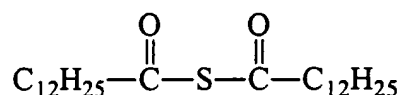
54. Stapfer [Ex. G] discloses thiocarboxylates at col. 11, lines 34-61. Dilauryl thiodipropionate is representative of Stapfer's class of thiocarboxylates and its structure is set forth below:



55. This compound, like other thiocarboxylates listed in Stapfer, is very different from the claimed reverse ester. A researcher in the PVH stabilization industry would have understood from Stapfer's representative compounds that Stapfer's thiocarboxylates had no free thiol groups.

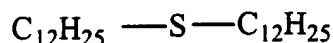
Furthermore, the sulfur atom of the Stapfer thiocarboxylates is found in the acid portion of the molecule, not the alkanol portion of the molecule as it is in reverse esters.

56. Stapfer [Ex. G] also discloses, as another class of non-tin compounds, thioanhydrides at col. 11, lines 62-68. Thiolauroic anhydride is representative of Stapfer's class of thioanhydrides and its structure is set forth below:



57. This compound, like other thioanhydrides, is very different from the claimed reverse esters because it has no free thiol (-SH) group and no ester group.

58. Finally, as the last class of non-tin compounds, Stapfer [Ex. G] discloses sulfides at col. 11, lines 69-73. Dilauryl sulfide is representative of this class of sulfur compounds:



59. This compound, like other sulfides, is very different from a reverse ester because it has no free thiol (-SH) group and it has no ester group.

60. Notwithstanding the broad list of compounds in the Stapfer patent, it does not include the claimed reverse esters, or the class of reverse esters generally. There is nothing in Stapfer that discloses or suggests the claimed reverse esters.

61. Stapfer provides absolutely no teaching about which possible combinations among the listed compounds exhibit synergism. In fact, Stapfer specifically and expressly disclaims *any* teaching about the selection of compounds:

*This invention does not reside in the selection of the particular stabilizer used and the compounds listed above are intended to be exemplary. These [sic] skilled in the art are able to determine the particular stabilizer, or stabilizers most suitable for a particular resin formulation and use. After the selection of the desired stabilizer, the present invention is applied by combining therewith a synthetic silicate powder.*

[Ex. G, col. 12, lines 5-12 (emphasis added).]

62. In my opinion, Stapfer provides no direction as to how to select the compounds to use in stabilizer combinations. Stapfer provides no more than a vague comment that poor stabilizers could be used in synergistic combinations. Stapfer does not suggest combining organotin mercaptides (claimed organotin compounds) with any sulfur, non-tin containing compounds. The PTO is thus simply wrong to state that "Stapfer directly discloses that [an organotin-sulfur compound] and a compound similar to [a reverse ester] may be combined to form a PVC stabilizer." See Defendant's Memo at page 15.

63. In my opinion, researchers would not have combined Stapfer with Gough. The Stapfer patent is directed to a solid support for stabilizers. The Gough patent is focused on organotin-borate compounds that are essentially useless. Further, the disclosure of a thiol co-stabilizer in Gough is tied to the organotin-borates. Gough provides no reason to use the co-stabilizer with any other organotin. To burrow into Gough to extract a reverse ester and combine it with the claimed organotin compounds, encompassed generically by Stapfer, would not be evident to researchers in the absence of the Chenard patent application telling them what to

combine. The Board was incorrect in assuming that the suggestion in Stapfer that poor stabilizers could be synergistically improved by the addition of other stabilizers somehow would have motivated researchers to combine the claimed components in the successful manner achieved by Drs. Chenard and Mendelsohn. [See Board Decision, Ex. N, at page 14.] Stapfer's disclosure of so many possible components without mentioning reverse esters confirms that without the Chenard invention, those in the PVH stabilization industry would not have thought of combining the claimed reverse esters with the claimed organotin compounds.

64. The Board's conclusion that "Stapfer discloses that a combination of organotin mercaptides and organic thio compounds produces synergistic results in stabilizing vinyl halide polymers" is also incorrect.<sup>7</sup> [Ex. O, Board's Decision on Rehearing, at page 3.] Stapfer gives no guidance about what specific combinations of compounds would exhibit synergism. Stapfer lists too many compounds for one of skill in the art to draw conclusions about which specific combinations of compounds might actually exhibit synergism, especially since such synergism is at best tangential to Stapfer's real invention: a silicate substrate.

65. The Board, referring to the Gough and Stapfer patents, incorrectly stated that "[a]ccordingly, based on these collective teachings, we find that one of ordinary skill in the art would have had a reasonable expectation of obtaining a synergistic effect when utilizing organic thiols and organotin mercaptides as stabilizers in vinyl halide polymers." [Ex. O, Board's Decision on Rehearing, at page 3.] I disagree. During the late 1970s, synergism was by no means expected or guaranteed when tin and sulfur compounds were combined. Nor is it expected or guaranteed today.

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<sup>7</sup> Note that a "thio compound" is simply a chemical which contains sulfur.

66. The import of the Board's findings is that all possible combinations of stabilizers exhibit synergism. [Ex. O, Board's Decision on Rehearing, at page 3]. This is incorrect. In my view, the Board did not appreciate the significance of evidence to the contrary, such as the data presented to the PTO in my 1982 Declaration. As explained below, for example, while synergism in some systems was observed in the testing reported in the 1982 Declaration, and in fact was unexpectedly enhanced for the claimed combinations, some non-claimed types of sulfur-containing compounds actually *decreased* the activity of the primary tin stabilizer (i.e., antisynergism).

#### **The Hechenbleikner '527 Patent**

67. Hechenbleikner '527 [Ex. H] teaches that mixtures of tin stabilizers with certain polymeric sulfur compounds can improve stabilization of halogen containing resins. [See Ex. H, col. 1, lines 27-34.] Hechenbleikner also teaches that those polymeric sulfur compounds can improve the lubrication properties of the resins for calendering and extrusion purposes. [See Ex. H, col. 4, lines 4-9.]

68. The polymeric sulfur compounds of Hechenbleikner are a unique class of sulfur compounds which are not within the scope of the claimed invention or similar to the claimed reverse esters. Hechenbleikner '527 would not have assisted those in the PVH stabilization industry to select the claimed reverse esters. Hechenbleikner's large, polymeric sulfur compounds appear to be useful because they contribute lubrication to the polymer. This disclosure of the alleged superiority of Hechenbleikner's unique sulfur compounds would focus the inquiry on those compounds and would therefore teach someone away from selecting a different class of sulfur compounds, such as the claimed reverse esters.

### **The Hechenbleikner '129 Patent**

69. Hechenbleikner '129 [ Ex. I] discloses combinations of tin stabilizers with mercapto *acid* esters, which are different from the claimed reverse esters. Hechenbleikner notes that sulfur compounds particularly useful for the invention have a thio linkage beta to a carbon atom having both a hydrogen atom and a carboxyl group attached to it, an arrangement which must necessarily be a mercapto acid ester. [See Ex. I, col. 17, line 73 - col. 18, line 43 (listing several suitable mercapto acid esters).]

70. This reference teaches the superiority of mercapto acid esters and therefore teaches someone skilled in the art away from selecting reverse esters. Gough's decision to test mercapto acid esters, but not the claimed reverse esters, reinforces the prevailing view at the time of the Chenard invention that mercapto acid esters were preferred thiol compounds.

### **The Weinberg Patent**

71. Weinberg [Ex. J] teaches the use of organotin-sulfur compounds as stabilizers. These are not used in combination with additional sulfur compounds. Weinberg does not teach, suggest, or motivate a researcher to arrive at the claimed invention.

### **The Kauder Patent**

72. Kauder [Ex. K] teaches a unique class of organotin-sulfur stabilizers for vinyl halide polymers. In addition, Kauder teaches synergistic combinations employing two organotin stabilizers. [See Ex. K, col. 6, lines 27-26.] Kauder does not teach, suggest, or motivate researchers to arrive at the claimed invention.



### **The Wowk Patent**

73. The Wowk patent [Ex. L] discloses the use of organotin stabilizers in which the tin is connected to both a sulfur atom and a halogen atom. Like Kauder, Wowk teaches that *tin* stabilizers may be used in combinations. [See Ex. L, col. 6, lines 28-col. 7, line 43.] Wowk does not provide any teaching, suggestion, or motivation to combine the claimed organotin compounds and the claimed reverse esters.

### **The Schroeder Patent**

74. Like Wowk, Schroeder [Ex. M] teaches organotin stabilizers in which the tin is connected to both a sulfur atom and a halogen atom. [See Ex. M, col. 1, lines 65-72.] I dispute the Defendant's statement that "Schroeder also teaches that [an organotin-halogen] compound can be combined with other known stabilizers including organic sulfur-containing compounds (such as [a claimed reverse ester])." [See Defendant's Memo, at pages 9-10.] Rather, Schroeder teaches, consistently with the Kauder and Wowk patents, that "[i]t is frequently desirable to incorporate into the polymer other organo-*tin* stabilizers in addition to the stabilizer according to the present invention . . . ." Col. 2, lines 18-20 (emphasis added). The addition of other *tin* stabilizers would be expected to increase stabilization, because workers in the PVC stabilization industry understood that overall tin content ordinarily is a good indicator of stabilization performance. Schroeder, however, does not teach, suggest, or motivate researchers to add sulfur compounds, let alone reverse esters, instead of other organotin stabilizers.

75. As is evident from the chart that appears in the Defendant's Memo, the *only* reference the PTO has cited that discloses the use of a reverse ester as a PVH co-stabilizer is the Gough patent. Despite the number and breadth of PVH stabilization references collected by the

PTO, the reverse ester was not a prominent feature of the PVH stabilization landscape at all. Even the Gough patent fails to provide specific test data or information with respect to the reverse ester's stabilization properties. There is nothing in any of the references, whether taken individually or collectively, to point in the direction of the narrow, specific class of claimed reverse esters for use as co-stabilizers with the claimed organotin compounds.

**IV. COMMENTS SUPPLEMENTAL TO REPRESENTATIVE EXAMPLES IN THE SPECIFICATION OF U.S. APPLICATION NO. 06/254,313, FILED APRIL 15, 1981 (Ex. D)<sup>8</sup>**

76. As noted above, one of the most significant aspects of the claimed invention is in its ability to decrease tin content and therefore cost. The results of testing show that the use of a claimed reverse ester in combination with a *low* amount of a claimed organotin compound can yield the same level of stabilization as a *larger* amount of claimed organotin compound alone.

77. Results in the specification demonstrate the practical ability of the claimed invention to reduce the amount of tin needed for stabilizing vinyl halide polymers. In the specification, the applicants estimated that the claimed invention allows the tin content to be reduced by 20% to 90%. [Ex. D, '759 application, page 20, line 3.] More generally, applicants estimate that there can be used one-third less metal with the claimed combinations. [Ex. D, page 20, lines 6-9.] Based on my experience, these estimates are, in my opinion, accurate.

78. When a polymer such as PVC degrades, the degradation is visually discernible, usually noticed by a loss of white color. Samples usually yellow first and eventually turn black.

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<sup>8</sup> The current application is Serial No. 07/870,759, which is a continuation application in a series beginning with 06/254,313. These two applications share the same written description portion of the specification.

One of the parameters used in the specification, as in the PVC industry, to measure the effectiveness of a stabilizer is the whiteness index which provides a quantitative measurement of color. I explain the significance of representative examples using whiteness index values in the specification below.

79. Some tests in the specification were conducted dynamically to show the degree of discoloration of the claimed invention as a function of time.

80. Other tests reported in the specification compared the resulting color of different stabilizer combinations both within and outside the scope of the claims of the present invention. These color results are given in whiteness index and yellowness index units, as measured by a colorimeter. These measurements are commonly used in the PVH stabilization industry as an objective means to compare the stabilizing effect of different PVH stabilizer compositions. A very white sample will have a high whiteness index value of about 100; values below this represent varying degrees of deviation from a perfect white. Yellowness index values, on the other hand, increase as more yellow appears within the degrading polymer. In general, as the whiteness index value increases, the yellowness index value decreases, and vice versa. Thus, a high whiteness index and low yellowness index indicate a desirably high degree of PVH stabilization.

81. Still other tests in the specification measured the melt viscosities of claimed compositions. Increased viscosity is another indication of PVH degradation. These viscosity tests therefore show the ability of the claimed invention to stabilize PVC to a degree that allows for a reduction in the amount of tin needed for effective stabilization. The results of these viscosity tests are presented in graph form at the end of the specification. [See Ex. D.]

1. **Example IV (pages 24-25)**

82. This example demonstrates that the compositions of the claimed invention achieve stabilization similar to that of conventional organotin compounds even with a significant reduction in tin content. A claimed organotin-sulfur stabilizer (the so-called "condensation product of butyl stannic acid and butyl thiostannic acid") was tested in combination with a claimed reverse ester, 2-mercaptoethyl stearate ("MES"). Five different stabilizer formulations were tested. The different formulations varied in amounts of organotin compound and MES used. The results are reported in the specification as follows:

83.

Table II<sup>9</sup>

	<u>% Tin Compound</u>	<u>%MES</u>	<u>Initial (minutes)</u>	<u>Browning</u>
A.	0.1	-	8'	12'
B.	0.1	1.00	16'	25'
C.	0.02	-	4'	6'
D.	0.02	1.00	7'	12'
E.	-	1.20	immediate	3'

84. The percent of tin compound is a measure of the percent by weight of organotin compound used. The percent of MES is a measure of the percent by weight of MES used. The "Initial (minutes)" entry measures the time it takes for discoloration to initially occur. "Browning" is a measure of the amount of time it takes for the PVH to reach a certain level of brown discoloration. Formulations A and C contain organotin compound but no MES;

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<sup>9</sup> All of the tables in my discussion of the Examples in the '759 application are reproduced from the patent specification; descriptive labels are added for clarification.

formulation E contains MES but no organotin compound. These three tests were used as controls.

85. The claimed combination, when the tin compound was only 0.02% (formulation D), produced almost the same heat stability, as measured by discoloration over time, as 0.1% of the tin compound without any MES (formulation A). Although the claimed formulation D showed initial discoloration one minute sooner (7') than the tin compound alone at a higher concentration (8'; formulation A), both formulations began browning at 12 minutes and thus exhibit similar longer-term stability. Thus, use of the reverse ester in formulation D allowed for a five-fold reduction in the amount of organotin used (0.02% versus 0.1% for formulations A and D, respectively) without any loss in stabilization efficacy.

86. The addition of the claimed reverse ester (1.00%) to the higher concentration of tin compound (0.1%) (formulation B) shows a dramatic, unexpected improvement to stability; approximately twice as much time passed, relative to formulations A or D, before any initial degradation (16') or browning (25') occurred.

87. In addition, applicants tested MES alone, which showed minimal stabilization activity of its own. Thus, the observed increase in stabilization is not due to an independent effect by the MES. The observed increase in stabilization must be synergistic; that is, the combined effect is greater than the contribution from each of the individual components.

## **2. Example VII (pages 26-29), Figures 1-2**

88. Torque rheometer curves, like those in Figures 1-4 [Ex. D, final pages] show the fusion time, heat stability and melt viscosity of a polymer composition. In general, better stabilizers exhibit a relatively constant torque value for a considerably longer period of time.

Figure 1 illustrates the principle that PVH stabilization is generally proportional to the percent tin content in the stabilizer composition. The stabilizer used in this experiment is a mixture of 0.5 moles butylthiostannoic acid and 1.5 moles di-n-butyltin-S,S'-bis(isodecylmercaptoacetate). Both of these compounds are claimed organotin-sulfur compounds. No reverse ester was included, however. In Figure 1, curve 1 represents a composition containing 1.8% of the stabilizer combination. Curve 2 represents a composition containing only 0.9% stabilizer combination. Curve 1 remains at a relatively constant value for much longer than curve 2. Thus, the higher the tin content, the more effective the stabilizer formulation. Recall that neither of these formulations (results recorded in curves 1 and 2) contain the claimed reverse ester.

89. For the experiment corresponding to Figure 2, MES was tested in combination with the tin compounds of Figure 1. Although they consist of one reverse ester and two organotin compounds, these combinations are nevertheless within the scope of the claimed invention. The following formulations were tested:

	<u>Tin Content (%)</u>	<u>MES (%)</u>
Curve 3	0.90	1.0
Curve 4	0.90	2.0
Curve 5	0.45	2.0

90. In Figure 2, it is seen that curve 4 outperforms the other two compositions, even though curve 4 has the same level of tin as curve 3. The two-fold increase of the claimed reverse ester unexpectedly increased the stability of the system without any increase in tin. The claimed reverse ester compounds were not known to be co-stabilizers with organotin-sulfur compounds, nor would they have been expected to give such strongly synergistic stabilization effects.

91. Significantly, curves 3 and 5 are approximately equivalent. This demonstrates that a formulation with reduced tin content and increased MES (curve 5) provides approximately the same level of stabilization as the higher tin/lower MES formulation (curve 3). This would have been unexpected by workers in the PVH stabilization field in the 1970s because it was generally believed that any reduction in tin content would diminish stabilization performance. The claimed invention thus allows the tin content to be reduced by half with approximately equivalent results.

3. **Example VIII (page 29, Figure 3)**

92. The claimed organotin compound used in these experiments is di-n-butyltin-S,S' bis(isooctyl mercaptoacetate). MES was added as the claimed reverse ester. These compounds were tested on a torque rheometer in the following amounts:

	<u>% Tin Content</u>	<u>% MES</u>
Curve 6	2.2	-
Curve 7	1.1	1

93. Curves 6 and 7, when examined in conjunction with curves 3 through 5, further demonstrate the unexpected synergism of the claimed combinations. Thus, MES combined with a different claimed organotin compound (*i.e.*, it differs from the combination of organotin compounds used in Figures 3 through 5) also yields superior stabilization effects.

94. Curves 6 and 7 in Figure 3 also demonstrate that nearly identical stabilization performance is observed for the claimed stabilizer composition (curve 7) compared to the use of twice as much tin stabilizer alone (curve 6).

4. **Example IX (page 30, Figure 4)**

95. In Example IX, the claimed organotin compound is a “condensation product of butylstannoic acid.” This organotin compound is combined with a claimed reverse ester, MES. These compounds were tested on a torque rheometer in the following amounts:

	<u>% Tin Content</u>	<u>% MES</u>
Curve 8	0.37	-
Curve 9	0.37	1
Curve 10	0.19	2

96. As shown in Figure 4, the curve 10 formulation significantly outperforms the other stabilizer compositions. This is very surprising because the composition of curve 10 has only half the tin content as either curve 8 or 9, which cuts against the generally accepted principle that stabilization ability is proportional to tin content (*see* curves in Figure 1).

97. Curve 8, the tin stabilizer compound alone, provides the weakest stabilization. The addition of 1% MES significantly improves its performance (curve 9). The addition of 2% MES even with a significant reduction in tin content dramatically improves its performance (curve 10).

5. **Example XIII (pages 35-36)**

98. Example XIII [Ex. D] demonstrates the use of the claimed invention with respect to the organotin-halide class of organotin compounds, which is the subject of pending claims 176-183 and 296-323. The combination of a claimed organotin-halide compound (butyltin trichloride), a claimed organotin-sulfur compound (the so-called “reaction product of butyltin trichloride, isooctyl mercaptoacetate and sodium sulfide in equimolar amounts”), and a claimed



reverse ester (mercapto ethyl oleate) was, in my opinion, a surprisingly good stabilizer combination.

6. Example XIV (pages 37-39)

99. Example XIV [Ex. D] demonstrates the unexpected nature of the superior synergistic co-stabilization effect of the claimed reverse ester. In my view, researchers in the PVH stabilization field would not have expected the reverse esters to be markedly superior co-stabilizers, compared to other sulfur-containing compounds, when combined with the claimed organotin compounds. This example demonstrates that various sulfur-containing compounds, having the same number of carbon atoms, are not equivalent in performance. The markedly superior results obtained with the claimed reverse esters would not have been expected by skilled workers in the PVH stabilization field during the late 1970s.

100. The sulfur compounds (each with twelve carbon atoms and thus with similar expected lubrication and volatility properties) tested were:

(1) lauryl mercaptan  $C_{12}H_{25}SH$

(a simple thiol *not within* the scope of the claimed invention);

(2) n-decylmercaptoacetate  $HSCH_2CO_2C_{10}H_{21}$

(a mercapto acid ester compound *not within* the scope of the claimed invention);

and

(3) 2-mercaptoethyl decanoate  $C_9H_{19}CO_2CH_2CH_2SH$

(a reverse ester *within* the scope of the claimed invention).

Each of these sulfur-containing chemicals was tested with 2 different claimed organotin-sulfur formulations: (1) monobutyltin (dodecylmercaptide) sulfide and (2) a combination of monobutyltin-(iso-octylmercaptoacetate)(sulfide) and butyltin trichloride.

**a. Results for Monobutyltin(dodecylmercaptide) Sulfide Plus Sulfur-Containing Compounds**

101. The claimed organotin-sulfur compound, monobutyltin (dodecylmercaptide) sulfide, was tested alone (formulation A) and then with each of the three sulfur compounds listed in ¶ 100 (formulations B, C, and D, respectively), but at a four-fold reduction of monobutyltin (dodecylmercaptide) sulfide (relative to formulation A), hence reduced tin content.

	<u>% Tin Compound</u>	<u>Sulfur Compound</u>	<u>% Sulfur Compound</u>	<u>W.I.</u>	<u>Y.I.</u>
A.	0.4	-	-	54.9	8.6
B.	0.1	(1)	0.3	34.6	15.8
C.	0.1	(2)	0.3	24.7	19.2
D.	0.1	(3)	0.3	57.8	7.7

102. Even at one-fourth the tin content, the stabilizer mixture containing the reverse ester of the claimed invention, 2-mercaptoethyl decanoate (formulation D), showed superior whiteness index and yellowness index values over the use of the tin compound alone (57.8 - 54.9 = 2.9 units improvement for whiteness; 8.6 - 7.7 = 0.9 units improvement for yellowness). In my opinion, these results are very unexpected because of the generally understood correlation between tin content and stabilization performance.

103. In addition, the claimed combination of reverse ester with a low level of monobutyltin (dodecylmercaptide) sulfide (formulation D) produces far better stability than the

other combinations containing non-claimed sulfur compounds (formulations B and C) at the same low level of monobutyltin (dodecylmercaptide) sulfide ( $57.8 - 34.6 = 23.2$  units improvement over lauryl mercaptan;  $57.8 - 24.7 = 33.1$  units improvement over n-decylmercaptoacetate). This is a remarkable result, demonstrating the unexpected superiority of the claimed combination. The yellowness index values confirm these results; for each of the sulfur compounds outside the scope of the claims, the values are significantly higher. In my opinion, these remarkably different results are very unexpected given the similarities (i.e. same number of carbon atoms) between the sulfur compounds.

**b. Results for Combination of Monobutyltin-(iso-octylmercaptoacetate) Sulfide and Butyltin Trichloride**

104. Here, the combination of monobutyltin-(iso-octylmercaptoacetate) sulfide and butyltin trichloride was tested alone (formulation A) and in combination with lauryl mercaptan, -decylmercaptoacetate, and 2-mercaptoethyl decanoate (formulations B, C, and D, respectively). Formulation D falls within the scope of the pending claims; the other formulations do not. The results are reported in the patent specification as follows:

	<u>% Tin Compound</u>	<u>Sulfur Compound</u>	<u>% Sulfur Compound</u>	<u>W.I.</u>	<u>Y.I.</u>
A.	0.4	-	-	37.7	14.7
B.	0.2	(1)	0.2	20.4	21.2
C.	0.2	(2)	0.2	3.4	27.9
D.	0.2	(3)	0.2	43.2	12.8

105. The claimed combination (formulation D) produced superior whiteness index results (43.2) to the organotin stabilizer alone (formulation A) (37.7), even when the amount of organotin compound stabilizer is reduced by half.

106. Furthermore, the results for the claimed combination are superior to the results for other organic sulfur compounds for a given level of organotin compound (0.2%). The yellowness index values are also superior for the claimed combination (12.8 with a reverse ester is much lower than the values above 20 for the other sulfur compounds). This stabilizer, as in Example XIII, also demonstrates the superiority of the invention for tin stabilizers in which the tin is bonded to halogen.

107. Moreover, the results for Examples XIV (a) and (b) demonstrate that the difference in stabilization performance is necessarily attributable to the claimed reverse ester, which has the -SH group in a different position relative to the ester (-CO<sub>2</sub>-) group in the molecule. The only significant structural difference between the mercapto acid ester (n-decyl mercaptoacetate) and the reverse ester (2-mercaptoethyl decanoate) is the position of the -SH group relative to the ester (-CO<sub>2</sub>-) group. Nothing in the prior art suggested such a dramatic improvement in stabilization performance by using a reverse ester in combination with an organotin stabilizer of the claimed invention.

108. In my opinion, the tests results described above (reported in the specification) demonstrate the remarkable properties of the Chenard invention. In my view, such properties would not have been expected and could not have been predicted at the time the invention was made.

## V. COMMENTS SUPPLEMENTAL TO 1982 DECLARATION

109. Several experiments that I conducted in support of the pending application are explained below, along with my comments referring to and clarifying my original 1982 Declaration [Ex. P], which I understand was submitted during prosecution of an earlier patent application that I understand is in the chain of related applications leading to the current application. I also describe how specific comparisons between sulfur compounds demonstrate the superiority of the combinations of the claimed invention. The Board did not appreciate that the only factor that could account for the differences in stabilization effects observed is the position of the -SH group relative to the ester (-CO<sub>2</sub>-) group in the claimed reverse esters (as opposed to other sulfur-containing compounds). Moreover, the Board did not appreciate the unpredictability of the results obtained with the claimed invention, nor did it appreciate the practical significance of the whiteness index results.

110. At the time of the 1982 Declaration, I understood that the major basis of the Examiner's rejection was the Gough and Stapfer<sup>10</sup> references. In connection with my 1982 Declaration, I had selected the following sulfur compounds from Examples 20 to 27 of Gough (Table III page 13 of U.S. 3,928,285):

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<sup>10</sup> I understand that at the time of the 1982 Declaration, the Examiner cited a different Stapfer reference against the application. [See Ex. T, Stapfer et al., *Organic Stabilizers for PVC Processing*, SPE JOURNAL 26: 22-26 (1972).] I am familiar with this article. In this reference, Stapfer strongly emphasized the use of thiolaureic anhydride, a compound far outside the scope of the pending claims. Thiolaureic anhydride is also one of the many compounds listed in the Stapfer '751 patent that is currently cited against the application. [Ex. G, col. 11, lines 65-66.]

isooctyl-thioglycolate  
isooctyl  $\beta$ -mercaptopropionate  
octadecylthioglycolate  
n-butylthioglycolate  
dodecylmercaptan  
benzylthioglycolate<sup>11</sup>  
octylmercaptan

From Stapfer, I had selected the following representative compound:

thiolauric anhydride

I had also tested the additional sulfur-containing compounds listed below which fall outside the scope of the pending claims to provide further comparisons between the claimed invention and other classes of sulfur compounds.

n-decylmercaptoacetate  
cyclohexylmercaptan  
diisooctylthiodipropionate

From the then-pending application, I had selected the following claimed reverse esters as representative:

mercaptoethyl tallate (oleate/linoleate)  
mercaptoethyl pelargonate (C<sub>9</sub>)  
mercaptoethyl ester of C<sub>108</sub> fatty acid  
mercaptoethyl stearate  
mercaptoethyl succinate<sup>12</sup>

111. The organotin-sulfur stabilizers had been selected to demonstrate the claimed invention with a variety of different compounds to support the scope of the claims. I had also

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<sup>11</sup> This sulfur compound became available only toward the end of the study and was not used in all of the experiments.

<sup>12</sup> At the time I tested these compounds, mercaptoethyl succinate was a reverse ester within the scope of the then-pending claims. The current pending claims, however, are directed to "mercapto alkanol esters of monocarboxylic acids," which would exclude mercaptoethyl succinate, an ester of a dicarboxylic acid. I am not aware of the use, as co-stabilizers, of any such reverse esters of dicarboxylic acids disclosed in the prior art.

tested an organotin-borate compound disclosed by Gough. The tin compounds were generally tested at 0.15 phr (parts per hundred parts resin), although in Experiment 3 I used 0.10 phr because the tin compound had a very high percentage of tin. Each tin compound has its own unique percentage of tin (calculations are provided). In general, tin compounds with similar percentages of tin which are used in equal amounts exhibit similar levels of stabilization. Experiments 4, 5, and 6 have nearly equivalent percentages of tin (about 30%) and would be expected to exhibit similar performance.

112. I will explain how particular sulfur compounds, when combined with claimed organotin compounds, demonstrate the stabilization effect of a single variable unique to the claimed reverse esters: the position of the -SH group in the molecule. The first tests I ran in Experiment 1 of my 1982 Declaration were run with chemicals available in the laboratory; later tests were performed to provide additional comparisons between similar compounds within and outside the claims.

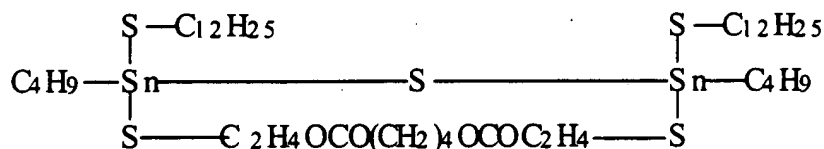
113. The results are given in terms of whiteness index values, values which, as explained above, are an accepted industrial measure of polymer degradation due to discoloration from a perfect white. The higher the whiteness index, the better stabilization. In evaluating the test results, I considered a difference of 3 units to be important; this degree of difference indicates that the results are not equivalent. A difference of 3 units is visually discernible and thus commercially significant, as discoloration is directly related to thermal stability. A high whiteness index value also indicates that the amount of stabilizer yielding the higher value may be reduced while still maintaining acceptable stabilization performance. Thus, as demonstrated by examples in the specification (*see* Section IV of this Declaration), a high whiteness index

value allows a corresponding reduction in the amount of tin stabilizer required, which means lower cost. I considered a difference of 5 or more units to be even more significant; this degree of difference indicates a substantial superiority of one formulation over another and a substantial opportunity to reduce the amount of tin needed for effective stabilization.

## EXAMPLES IN THE DECLARATION IN SUPPORT OF PATENTABILITY

### 1. Experiment 1

114. In this experiment, I used a claimed organotin compound that can be chemically represented as follows:



In order to compare test results on a *tin* content basis, I calculated the percent of tin within the organotin compound used; in this experiment the tin content of the organotin compound was 22.1%. The tin compound above was tested by itself and then in combination with each of the listed sulfur compounds, respectively:



<u>STABILIZERS</u>			<u>WI</u>
A. <sup>13</sup>	tin stabilizer alone		50.2
B.	diisooctylthiodipropionate	+ tin stabilizer	51.9
C.	n-decylmercaptoacetate	+ tin stabilizer	27.3
D.	cyclohexylmercaptan	+ tin stabilizer	41.2
E.	thiolauric anhydride	+ tin stabilizer	29.4
F.	mercaptoethyl tallate	+ tin stabilizer	56.0

115. As the results show, the tin stabilizer alone gave very good stabilization (high whiteness index value of 50.2). Formulations (C), (D) and (E), which are not within the scope of the claimed invention, exhibit *anti-synergistic* behavior -- i.e., the addition of a sulfur compound actually *worsens* the performance of the organotin stabilizer and results in a lower, less desirable whiteness index value. Despite the anti-synergism of some combinations with this tin stabilizer, the claimed reverse ester (F) improves the organotin compound to a significant degree (more than 5 whiteness index units). Combination B exhibits only minimal improvement (1.7 units) over the organotin compound alone (A).

116. A comparison of the results for formulations C (27.3) and F (56.0) is surprising because the sulfur compound in C is a mercapto acid ester. As discussed earlier in this Declaration, mercapto acid esters have an apparently minor structural difference in the position of the -SH group relative to the ester (-CO<sub>2</sub>-) group in the molecule. Although the sulfur compounds n-decylmercaptoacetate and mercaptoethyl tallate are structurally different, the two sulfur compounds possess similar functional groups. Nothing in the prior art taught that they would exhibit remarkably different stabilization performance. The difference of (56.0-27.3) **28.7** whiteness index units between these compounds is unexpected.

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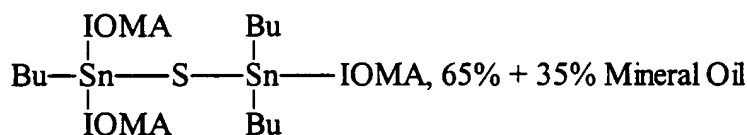
<sup>13</sup> Please note that the examples have been re-labeled with capital letters (A through F) for purposes of discussion.

117. Although the Board asserted that sulfur compounds and organotin stabilizers were synergistic, and known to be so, my data clearly demonstrates that this was not always the case. [See Board's Decision, Ex. N, at pages 13-14].

118. These results demonstrate the unpredictability of stabilizer combinations: combinations of the various classes of stabilizers can (1) exhibit synergism; (2) exhibit stabilization equal to the sum of the effect of independent stabilizers; (3) show no improvement in combination; or (4) exhibit antisynergism. One could not have had a reasonable expectation of success for all combinations of organotin-sulfur and organotin-halogen compounds with other sulfur compounds, including the claimed reverse esters.

## 2. Experiment 2

119. In this experiment, the following organotin compound, containing 14.5% of tin, was tested:



IOMA = isooctylmercaptoacetate, a sulfur-containing organic group;  
Bu = butyl, a non-sulfur-containing organic group

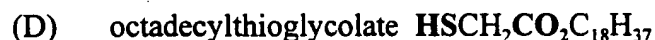
The organotin compound above was tested by itself (A) and then in combination with each of the listed sulfur compounds (B-K, respectively). Formulation (J) falls within the scope of the claims. I carefully selected the sulfur compounds to demonstrate the effect of particular variables.

STABILIZERSWI

A.	tin stabilizer alone		1.7
B.	i-octylmercaptoacetate	+ tin stabilizer	0.4
C.	i-octyl $\beta$ -mercaptopropionate	+ tin stabilizer	3.7
D.	<b>octadecylthioglycolate</b>	+ tin stabilizer	<b>2.3</b>
E.	n-butylthioglycolate	+ tin stabilizer	7.4
F.	dodecylmercaptan	+ tin stabilizer	-8.7
G.	octylmercaptan	+ tin stabilizer	-6.5
H.	benzylthioglycolate	+ tin stabilizer	16.9
I.	thiolauric anhydride	+ tin stabilizer	16.9
J.	<b>mercaptoethyl stearate</b>	+ tin stabilizer	<b>22.9</b>
K.	mercaptoethyl succinate	+ tin stabilizer	22.1

120. In my opinion, the result obtained with a claimed combination of the organotin-sulfur compound and the reverse ester (J) is significantly superior to the results obtained with other sulfur compounds in combination with the organotin-sulfur compound (i.e., non-claimed combinations).

121. In particular, the results for formulations (D) and (J) (in bold type for discussion purposes) can only be explained by the position of the -SH group in the sulfur compound. Each of the sulfur compounds has the same number of carbon atoms. The primary difference between octadecylthioglycolate (D) and mercaptoethyl stearate (J) is the -SH (or "HS-") position relative to the ester (-CO<sub>2</sub>-) in the molecule; the claimed mercaptoethyl stearate has the -SH in what chemists call the "alkanol" portion of the compound, as shown in (J) below.

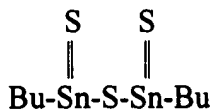


122. Persons working in the PVC stabilization field would not have expected that the position of the -SH group would produce the large differences in whiteness index values of **19.6** (22.9 - 2.3) units.

123. A comparison between formulations (B) and (D) (both outside the scope of the claims) further illustrates the unexpectedness of the results for (D) and (J). The sulfur compound of (B) has 10 carbon atoms, whereas the sulfur compound of (D) has 20; yet the two compounds exhibit relatively similar stabilization properties. The number of carbon atoms therefore does not appear to account for the difference in performance between these two compounds. Thus, any lubrication effect from the long chain of carbon atoms<sup>14</sup> cannot explain the outstanding performance of the sulfur compound in (J) because other similarly-long chain molecules do not exhibit its remarkable stabilization. In my opinion, given the fact that the same organotin compound was used in all these tests, the only possible variable that is responsible for the superior performance of (J) is the position of the -SH group in the molecule --i.e, the fact that the sulfur compound is a reverse ester-- something the prior art in no way taught, predicted or suggested.

### 3. Experiment 3

124. The organotin compound used in this experiment is called butylthiostannoic anhydride. Containing 53.1% tin, its chemical structure is as follows:



Bu = butyl, a non-sulfur-containing organic group

This tin compound, which is within the scope of the claimed invention, was tested by itself (A) and then in combination with each of the listed sulfur compounds (B-K, respectively).

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<sup>14</sup> In general, compounds with longer carbon chains are superior lubricants. It is often desirable to have lubricants in the PVH manufacturing process.

Formulation (J) is within the scope of the claimed invention. I used less of this tin stabilizer (0.10 phr vs. 0.15 phr for the other experiments) because of its inherently high tin content.

<u>STABILIZERS</u>			<u>WI</u>
A.	tin stabilizer alone		30.3
B.	i-octylthioglycolate	+ tin stabilizer	42.5
C.	i-octyl $\beta$ -mercapto propionate	+ tin stabilizer	49.2 <sup>15</sup>
<b>D.</b>	<b>octadecylthioglycolate</b>	+ tin stabilizer	<b>29.7</b>
E.	n-butylthioglycolate	+ tin stabilizer	46.7
F.	dodecylmercaptan	+ tin stabilizer	39.8
G.	octylmercaptan	+ tin stabilizer	42.9
H.	benzylthioglycolate	+ tin stabilizer	45.0
I.	thiolauric anhydride	+ tin stabilizer	44.5 <sup>16</sup>
<b>J.</b>	<b>mercaptoethyl stearate</b>	+ tin stabilizer	<b>50.4</b>
K.	mercaptoethyl succinate	+ tin stabilizer	55.6

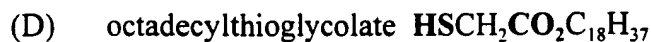
125. The results for the claimed combination of organotin compound and reverse ester (J) are significantly higher than those obtained with other sulfur compounds in combination with the same organotin compound. In particular, for the reasons stated for Experiment 2, the comparison between the sulfur compounds of (D) and (J) demonstrates that the only variable that can account for the performance of the claimed combination is the position of the -SH group relative to the ester (-CO<sub>2</sub>-) group in the sulfur-containing compound. Each of these compounds (D) and (J) has the same number of carbon atoms. The primary difference between octadecylthioglycolate (D) and mercaptoethyl stearate (J) is the -SH position relative to the ester (-CO<sub>2</sub>-) in the molecule; the mercaptoethyl stearate has the -SH in the alkanol portion of the

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<sup>15</sup> This value originally appeared in the 1982 Declaration as 49.5. This appears to have been a transcription error from my laboratory notebook. [See Ex. R, Lab Notebook No. 4573, at page 34]. The difference of  $\pm 0.3$  units, in my opinion, is insignificant. I report here the value appearing in my laboratory notebook.

<sup>16</sup> Brown Shade.

ester, as shown below, and is therefore a reverse ester. Octadecylthioglycolate (D), on the other hand, is *not* a reverse ester.



126. It was very surprising that this single difference of the position of the -SH group would produce the large difference in whiteness index values of 20.7 (50.4-29.7) units seen above. This difference is extremely surprising for such otherwise similar compounds.

#### 4. Experiment 4

127. This claimed organotin stabilizer, monobutyltin(isooctylmercaptoacetate)sulfide, which contains 29.0% tin, was tested:



This tin compound was tested alone (A) then in combination with each of the listed sulfur compounds (B-K). This commercially available tin stabilizer, within the scope of the claimed invention, was known to exhibit moderate stabilization performance. Again, formulation (J) is a claimed combination.

<u>STABILIZERS</u>			<u>WI</u>
A.	tin compound alone		27.7
B.	isooctyl thioglycolate	+ tin stabilizer	35.6
C.	isooctyl $\beta$ -mercaptopropionate	+ tin stabilizer	38.8
D.	<b>octadecylthioglycolate</b>	+ tin stabilizer	<b>29.0</b>
E.	n-butylthioglycolate	+ tin stabilizer	40.0
F.	dodecylmercaptan	+ tin stabilizer	28.4
G.	octylmercaptan	+ tin stabilizer	36.0
H.	benzylthioglycolate	+ tin stabilizer	49.7
I.	thiolauric anhydride	+ tin stabilizer	38.0

J.	mercaptoethyl stearate	+ tin stabilizer	54.8
K.	mercaptoethyl succinate	+ tin stabilizer	56.7

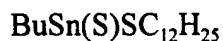
128. I consider the results obtained with the reverse ester in (J) to be significantly superior to those obtained with other sulfur compounds. As discussed for Experiments 2 and 3 above, the comparison between the sulfur compounds in (D) and (J) is especially striking because each of these compounds has the same number of carbon atoms. The primary difference between octadecylthioglycolate and mercaptoethyl stearate is the -SH position relative to the ester (-CO<sub>2</sub>-) in the molecule; the mercaptoethyl stearate has the -SH in the alkanol portion of the ester, and is thus a reverse ester, as shown below:

(D)	octadecylthioglycolate	$\text{HSCH}_2\text{CO}_2\text{C}_{18}\text{H}_{37}$
(J)	mercaptoethyl stearate	$\text{C}_{17}\text{H}_{35}\text{CO}_2\text{CH}_2\text{CH}_2\text{SH}$

129. It was very unexpected that this single difference of the position of the -SH group would produce a difference in whiteness index values of 27.8 (54.8 - 29.0) units. As explained above, the only possible variable that could be responsible for the superior performance of (J) is the position of the -SH group in the molecule, which puts it in the class of claimed reverse esters, something the prior art in no way taught, predicted or suggested.

## 5. Experiment 5

130. The following claimed organotin compound, monobutyltin (sulfide) dodecylmercaptide (29.1% tin) was tested:



This organotin compound, known to exhibit moderate stabilization activity, is within the scope of the claimed invention. It was tested by itself (A) and then in combination with each of

the listed sulfur compounds (B-K). These sulfur compounds were selected to provide additional comparisons to Experiments 1-4. Formulations (I) and (J) are claimed combinations.

<u>STABILIZERS</u>			<u>WI</u>
A.	tin compound alone		24.1
B.	isooctyl thioglycolate	+ tin stabilizer	39.9
C.	<b>isooctyl <math>\beta</math>-mercaptopropionate</b>	+ tin stabilizer	<b>39.7</b>
D.	octadecylthioglycolate	+ tin stabilizer	28.0
E.	n-butylthioglycolate	+ tin stabilizer	41.0
F.	dodecylmercaptan	+ tin stabilizer	25.9
G.	octylmercaptan	+ tin stabilizer	31.0
H.	thiolauric anhydride	+ tin stabilizer	29.2
I.	<b>mercaptoethyl pelargonate</b>	+ tin stabilizer	<b>64.4</b>
J.	<b>mercaptoethyl-C108 acid ester<sup>17</sup></b>	+ tin stabilizer	<b>59.9</b>

131. It is seen that the claimed reverse esters in (I) and (J) dramatically improve stabilization performance above other sulfur compounds. In particular, a comparison between the sulfur compounds of (C) and (I) dramatically illustrates the unexpected nature of the improvement (24.7 units difference) because these two compounds have the same number of carbon atoms, but the -SH group is in a different position in the molecules relative to the ester (-CO<sub>2</sub>-) group, as illustrated below:

(C) isooctyl  $\beta$ -mercaptopropionate       $C_8H_{17}O_2CCH_2CH_2SH$

(I) mercaptoethyl pelargonate       $HSCH_2CH_2O_2CC_8H_{17}$

Thus, the sulfur compound in (I) is a reverse ester; the sulfur compound in (C) is not.

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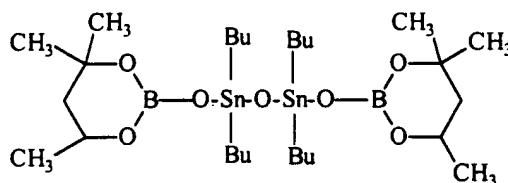
<sup>17</sup> C108 is a coconut-based mixed C<sub>8</sub>-C<sub>14</sub> fatty acid obtained from the Proctor & Gamble Corp.



Nothing in the prior art taught or suggested that a dramatic level of stabilization improvement could be obtained by this reversal of the position of the -SH group relative to the ester (-CO<sub>2</sub>-) group. This difference of 24.7 units is extremely surprising.

## 6. Experiment 6

132. In this experiment, I tested an organotin-borate that was disclosed as product a-1, of Example 1, in the Gough patent. It contains 31.0% tin, and has the following structure:



1, 3-bis(4, 4, 6-trimethyl-1, 3, 2-dioxaborinyl-2-oxy)-  
(tetra-n-butyl)distannoxane)

This organotin-borate is used in Examples 20 to 27 of the Gough patent (cols. 13-14, Table III). The limited availability of this product (lab sample) forced me to select only the sulfur compounds in Gough with the best stabilization results, as indicated by Gough in Table III. Thus, I had no reason to believe that a combination of Gough's organotin-borate with a reverse ester, let alone the one claimed reverse ester disclosed in Gough, would produce better results than any of the other combinations Gough explicitly preferred. The organotin-borate is significantly different from the claimed organotin-sulfur and organotin-halogen compounds because the tin (represented by the symbol "Sn") is bonded to oxygen, which is then bonded to boron (represented by the symbol "B"). There are no tin-sulfur or tin-halogen bonds in the Gough organotin-borates, unlike the claimed organotin compounds.

133. I tested the organotin-borate of Gough alone (A) and in combination with the following sulfur compounds (B-G), yielding whiteness index results as shown:

<u>STABILIZERS</u>			<u>WI</u>
A.	organotin-borate alone		deep purple/brown <sup>18</sup>
B.	i-octylthioglycolate	+ organotin-borate	-20.2
C.	i-octyl $\beta$ -mercaptopropionate	+ organotin-borate	5.4
D.	n-butylthioglycolate	+ organotin-borate	-7.5
E.	benzylthioglycolate	+ organotin-borate	10.9
F.	octylmercaptan	+ organotin-borate	-10.8
G.	thiolauric anhydride	+ organotin-borate	-25.9

134. The organotin-borate alone (A) has no stabilizing effect, as shown by the deep purple color, despite a tin content (31%) comparable to formulations used in Experiments 4 (29% tin) and 5 (29.1% tin) above. This is in full agreement with Gough's results (compare examples 12 and 27 of Gough), which reflect the failure of Gough's organotin-borate compounds. Given the extremely poor stabilization performance of the Gough organotin-borate, almost all of the tested compounds improve the performance of the Gough organotin-borate. The "improved" results obtained with combinations of organotin-borates and the above-listed sulfur compounds were not, in my opinion, good enough for commercial use.

135. Moreover, the "improvement" is not necessarily synergistic, contrary to Gough's assertions in col. 2, lines 13-16. In Table II of the Gough patent [Ex. F at cols. 13-14], the use of the thiol compound octylthioglycolate alone showed *better* stabilization than use of Gough's

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<sup>18</sup> This color was reported as deep purple in the 1982 Declaration, but as deep purple/brown in my laboratory notebook. [See Ex. R, Lab Notebook No. 4573, at page 35.] This discrepancy appears to be a transcription error and does not affect my analysis of the result. The sample was so badly degraded that no whiteness index value was calculated. Whiteness index values are not as accurate for such low sample readings. I report here the result appearing in my laboratory notebook.

organotin-borate compounds alone. [See Gough patent, Ex. F, cols. 13 and 14, Table II, Experiments 16-19.] The observed stabilization improvement for combinations of thiol compounds and organotin-borates may be due to the sum of their small independent contributions to stability. I would not characterize these systems as exhibiting synergism, which requires that the two components together demonstrate better results than the sum of their individual contributions.

136. The results I obtained using the organotin-borates confirms what researchers would have gleaned from the Gough patent itself: that the organotin-borate/thiol combinations of Gough were commercially useless. Indeed, in my tests, most of the claimed organotin-sulfur compounds *alone* exhibit stabilization effects superior to that of the organotin-borate and sulfur compound combinations tested. The teaching away in Gough from the organotin compounds (which are disparaged as unsatisfactory for a number of reasons, *see* Gough, Ex. F, col. 1, lines 44-66) and the inferior performance of Gough's combinations all tend to teach away from relying on any portion of Gough for the alleged teaching or suggestion to combine a reverse ester with a conventional organotin-sulfur or organotin-halogen stabilizer. As explained above, Gough provides no motivation whatsoever to continue experimenting with Gough's unsatisfactory compositions in myriad combinations.

137. On the basis of the experiments described above, the results obtained with the claimed compositions are superior to a substantial degree over those of the prior art; further, those results could not be predicted from the prior art.

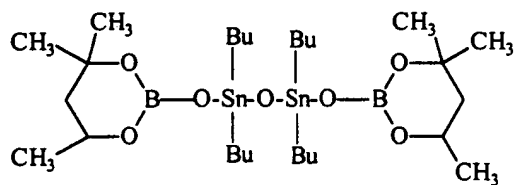
## VI. COMMENTS REGARDING THE NEW TESTING PERFORMED BY CHRISTOPHER A. BERTELO

138. I have reviewed new test data that has been obtained under the direction of Dr. Christopher A. Bertelo, comparing the claimed invention with a combination of Gough's organotin-borate and a reverse ester. I chose not to make this comparison in my original 1982 Declaration because the organotin-borate stabilizer was so poor, and the amount of organotin-borate available was very limited.

139. Although it is impossible to exactly replicate the reaction conditions of my experiments performed almost 20 years ago, the observed trends in Dr. Bertelo's experiments are comparable to the observed trends in my 1982 Declaration experiments. I understand from Dr. Bertelo that he used the same amounts of reagents and approximately the same conditions as those described in my 1982 Declaration. All organotin compounds were tested at 0.15 phr (parts per hundred parts resin). The sulfur compounds were tested at 0.3 phr.

140. Dr. Bertelo tested a Gough organotin-borate and two organotin-sulfur compounds within the scope of the claims:

- (1) Gough organotin-borate compound (outside the scope of the claimed invention):



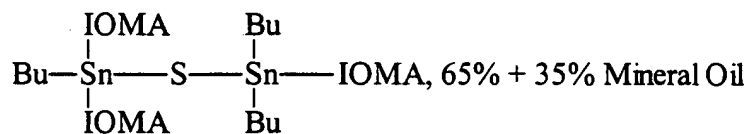
1,3-bis(4,4,6-trimethyl-1,3,2-dioxaborinyl-2-oxy)(tetra-n-butyl-distannoxane)

This organotin-borate, the product a-1 (Example 1) in Gough, is used in Examples 20 to 27 of Gough (cols. 13 and 14, Table III).

- (2) "T<sub>1</sub>": BuSn(S)SC<sub>12</sub>H<sub>25</sub> Monobutyltin (sulfide)(dodecylmercaptide)

This organotin-sulfur compound is within the scope of the claimed invention.

- (3) "T<sub>2</sub>":



IOMA = isooctylmercaptoacetate

This organotin-sulfur compound is within the scope of the claimed invention.

141. These organotin compounds were tested alone and then in combination with the three following sulfur compounds:

- (1) 2-thioethyl octanoate  $\text{CH}_3(\text{CH}_2)_6\text{CO}_2\text{CH}_2\text{CH}_2\text{SH}$

(a reverse ester within the scope of the claimed invention and set forth in Gough's specification at col. 8, line 64, but not tested by Gough, also called 2-mercaptoethyl octanoate)

- (2) 2-mercaptoethyl stearate  $\text{C}_{17}\text{H}_{35}\text{CO}_2\text{CH}_2\text{CH}_2\text{SH}$

(a reverse ester within the scope of the claimed invention)

- (3) octadecyl mercaptoacetate  $\text{HSCH}_2\text{CO}_2\text{C}_{18}\text{H}_{37}$

(a mercapto acid ester not within the scope of the claimed invention, also called octadecylthioglycolate)

142. The whiteness index results for combinations of organotin compounds and the sulfur compounds are reported in Table 1 below.

**TABLE 1**  
**Whiteness Index Values for Stabilizer Combinations**

Sulfur Compounds	Organotin-borate	T <sub>1</sub>	T <sub>2</sub> (½ tin content)
2-thioethyl octanoate	-9.5	<b>45.7*</b>	<b>2.1</b>
2-mercaptoethyl stearate	-13.7	<b>40.3</b>	<b>5.5</b>
octadecyl mercapto acetate	-22.6	9.2	-28.4

Note: The *higher* the number, the better the result.

\* Results in bold pertain to claimed combinations.

143. The data in Table 1 illustrates that all combinations of organotin-borates with sulfur compounds give negative whiteness index values, regardless of the identity of the sulfur compound. Even the claimed reverse ester, when combined with the Gough organotin-borate, did not produce acceptable results. These results would be considered unacceptable by anyone who wished to use these stabilizers. Combinations of the claimed invention, however, give results (shown in bold-face type) that are significantly superior to all combinations involving organotin-borates. The comparison between T<sub>1</sub> and the organotin-borate is especially striking because these tin compounds have approximately the same tin content (about 30%). Clearly, combinations of the claimed invention (whiteness index values of 45.7 and 40.3) are superior to combinations of organotin-borates with the same reverse esters, which yield negative whiteness index values of -9.5 and -13.7, respectively.

144. The comparison between  $T_2$  and the organotin-borate is even more striking, because  $T_2$  shows much better performance despite  $T_2$  having only about *half* the tin content (about 15%) of the organotin-borate (about 30%).  $T_2$  gives whiteness index values of 2.1 and 5.5, which are well above the negative whiteness index values of -9.5 and -13.7 for the organotin-borate with the same sulfur compounds. While the result obtained using the  $T_2$  system is not nearly as good as that obtained using the  $T_1$  system, relative to Gough's organotin-borate system,  $T_2$  is superior.

145. Combinations of the claimed invention are also superior to combinations of  $T_1$  and  $T_2$  with a sulfur compound outside the scope of the claims, such as octadecyl mercaptoacetate. The octadecyl mercaptoacetate gives terrible stabilization results when combined with both the organotin-borate (-22.6) and  $T_2$  (-28.4). Although the whiteness index value for this compound in combination with  $T_1$  is above zero (9.2), it is nowhere near as good as the results for the claimed sulfur compounds in combination with  $T_1$  (45.7 and 40.3).

146. As described earlier in this Declaration, these results can only be explained by the position of the -SH group in the molecule relative to the ester (-CO<sub>2</sub>-) group.

2-mercaptoethyl stearate       $C_{17}H_{35}CO_2CH_2CH_2SH$

octadecyl mercaptoacetate       $HSCH_2CO_2C_{18}H_{37}$

As the molecular formulas of these compounds show, the only significant difference between these compounds is the position of the -SH group in the compound. Each compound has 20 carbon atoms and a similar arrangement of atoms. It would have been completely unexpected that this mere positional difference in the compound would yield stabilization results as strikingly different as what is shown here.

147. Yellowness index values were also recorded for the 9 stabilizer combinations tested:

**TABLE 2**  
**Yellowness Index Values for Stabilizer Combinations**

Sulfur Compounds	Organotin-borate	T <sub>1</sub>	T <sub>2</sub> (½ tin content)
2-thioethyl octanoate	32.6	<b>13.4</b>	<b>29.1</b>
2-mercaptoethyl stearate	34.7	<b>15.3</b>	<b>27.7</b>
octadecyl mercapto acetate	39.1	26.1	42.4

Note: The *lower* the number the better the result.

148. The yellowness index values in Table 2 confirm the results of Table 1. Combinations of the claimed invention, which are shown in bold-face type, give results that are significantly superior to the combinations of organotin-borates. T<sub>1</sub> and the organotin-borate offer the best comparison because these tin compounds have approximately the same tin content (about 30%). As discussed above, tin content has always been the best general indicator of stabilization performance, with higher levels of tin usually providing better stability. Clearly, combinations of the claimed invention (much lower yellowness index values of 13.4 and 15.3 for T<sub>1</sub>) are superior to combinations of organotin-borates with the same sulfur compounds, which yield higher yellowness index values of 32.6 and 34.7.

149. Table 3 shows the whiteness and yellowness index values for the organotin-borate and the organotin-sulfur compounds used alone as controls.



**TABLE 3**  
**Whiteness and Yellowness Index Values for Stabilizer Control Experiments**

Controls	Organotin-borate	T <sub>1</sub>	T <sub>2</sub> (½ tin content)
Whiteness Index	-20.1	19.1	-18.9
Yellowness Index	59.9	23.4	41.4

150. It is readily apparent that the organotin-borate is an extremely poor stabilizer, both by itself (-20.1 whiteness index value) and, as we have demonstrated above, even in combination with reverse esters or any other type of sulfur-containing compound (all such combinations yield only negative whiteness index values). Even if Gough had tested reverse esters, his results would not have motivated a person of ordinary skill in the art to select reverse esters from among the infinite number of organic thiols disclosed in Gough and combine them with the claimed organotin compounds. The combined results simply do not provide any motivation to do any further work using anything taught by Gough. Organotin-borates do not work well, even in combination with Gough's sulfur compounds.

## **VII. CONCLUSION**

151. On the basis of the experiments described above, it is my opinion that the results obtained with the compositions of the claimed invention are substantially superior to combinations of organotin-borates with reverse esters.

152. These results also confirm the general trends I observed almost twenty years ago about the superiority of claimed combinations of organotins and reverse esters as compared to combinations of organotins with other sulfur compounds.

153. In view of the above, it is my opinion that the prior art does not teach, suggest, or motivate one skilled in the art to arrive at the claimed combinations of stabilizers for vinyl halide polymers.

I declare under penalty of perjury that the foregoing is true and correct.

Executed on: May 4, 2000

  
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MICHEL FOURE